Incommensurate Charge Density Wave as Quantum Space-Time Crystal

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A quantum time crystal (QTC) is a novel quantum mechanical ground state that was recently proposed by Wilczek. Although many QTC models have been proposed, it is not clear yet if such states are possible. We propose the idea that an incommensurate charge density wave (ICDW) forms a quantum space crystal (QSC), which is an extension of a QTC in the spatial domain. Consequently, a rotating ring-shaped ICDW naturally forms a quantum space-time crystal (QSTC), which combines the two concepts of QTC and QSC. The breaking of space-time translation symmetry is understood using time-dependent density matrices. Furthermore, we show that this model can be observed in real systems such as TaS_3 ring crystals at finite temperature. Our results suggest that QTC/QSTC can exist.

INTRODUCTION

Wilczek recently proposed that certain quantum systems, called quantum time crystals (QTC), can spontaneously break continuous time translation symmetry by forming a time-periodic structure in their ground state [1]. A QTC, unlike conventional spatial crystals, exhibits off-diagonal long-range order (ODLRO)[2]. In other words, the time translation symmetry of an observed QTC can be broken by the presence of a periodic lattice in time, but an unobserved one is a superposition of such lattices that is temporally uniform [3]. This means that observation breaks time translation symmetry. There is a similar proposition in spatial coordinate, namely, supersolid [4], which we call a quantum space crystal (QSC) in this paper because a supersolid is a spatial crystal with ODLRO. That is, the ground state of an unobserved QSC is a uniform superposition of lattices in space, but observing the system breaks the space-translation symmetry because of the presence of a lattice. QSC and QTC are two important predictions that are related to the observation problem, and they have to be verified experimentally.

Wilczek's original proposition is that QTCs are formed by the spontaneous breaking of time translation symmetry, but Bruno [5] and Watanabe and Oshikawa [6] proved that this scenario is not possible in the infinite volume limit. However, these criticisms do not remove the possibility that QTCs can be formed in finite systems where time translation symmetry breaking does not result from spontaneous symmetry breaking, which is what we will discuss in this paper. In fact, we will show that our model agrees with Bruno's result for the infinite volume limit but Bruno's result does not apply for a finite system. We will also show that our model is a counter example to Watanabe and Oshikawa's result.

We propose that a ring-shaped QSC rotating in the ground state ($T = 0$) forms a quantum space-time crystal (QSTC), which combines the two concepts of QTC and QSC. A good candidate for a QSC is an incommensurate charge density wave (ICDW) [7, 8]. A charge density wave (CDW) is a well known phenomenon that spontaneously breaks space translation symmetry in quasi-one-dimensional systems [8]. It is a periodic modulation of the electric charge density that occurs due to electron-phonon coupling. If the ratio of the CDW wavelength λ and the lattice constant a of the crystal is close to an irrational number, then the CDW is said to be incommensurate. Incommensurability is the most important feature/concept that is not present in other QTC models that are similar to ours, such as the space-time crystal model proposed by Li *et al.* [9] or the Cooper pair density wave model proposed by Nozières [10]. The sliding of an ICDW is described by a gapless Nambu-Goldstone (phason) mode [8]. Consequantly, the energy of an ICDW is independent of its phase (i.e. position) [8], which implies that the expected ground state of an ICDW ring is a superposition of ICDWs with different phases. Moreover, several models [11–15] show that an ICDW ring can couple to electromagnetic potentials and slide without energy loss. We first calculate the quantum mechanical wave function of an ICDW formed on a ring-shaped quasi-one dimensional crystal. We then show that an ICDW ring threaded by a magnetic flux rotates in the ground state and form a QSTC. Finally, we discuss some experimental realizations at finite temperature and discuss how our model avoids the criticisms of time crystals.

QUANTUM WAVEFUNCTION OF ICDW RINGS

It is well known that the electric charge density of a quasi-one-dimensional crystal becomes periodic by opening a gap at the Fermi wavevector $Q = \pm k_\mathrm{F}$, hence forming a CDW state with a wavelength $\lambda = \pi/k_\mathrm{F}$ [8]. Consider a CDW formed on an ideal ring-shaped quasi-one-dimensional crystal with radius R. The order parameter that describes this CDW ring is a complex scalar $\Delta(x,t) = |\Delta(x,t)| \exp[i\xi(x,t)]$, where $|\Delta(x,t)|$ is the size of the gap at $Q = \pm k_\mathrm{F}$, $\xi(x,t)$ is the phase of the CDW, $x \in [0, 2\pi R)$ is the coordinate on the crystal, and t is the time coordinate. The charge density is

$$\rho_\mathrm{CDW}(x,t) = \rho_0 + \rho_1 \cos(2k_F x + \xi(x,t)) \tag{1}$$

where ρ_0 is the average charge density and ρ_1 is the amplitude of the wave. If the CDW is incommensurate, it can rotate freely without energy loss thanks to translation invariance, so the dynamics of the ICDW is understood by the phase ξ. We further assume a rigid-body model of ICDW, i.e. the ICDW ring does not deform locally and the phase $\xi(x,t) = \xi(t)$ does not depend on position. We introduce the collective coordinate $X(t) = \xi(t)/2k_\mathrm{F}$, which moves together with the ICDW. If the ICDW ring with radius R has N crests, then the wavelength λ of the ICDW is related to k_F, N and R by

$$2k_F = \frac{2\pi}{\lambda} = \frac{N}{R}. \tag{2}$$

Therefore, from the definition of the collective coordinate we obtain

$$\frac{\partial N\phi}{\partial t} = \frac{\partial \xi(t)}{\partial t} \tag{3}$$

where $\phi = \phi(t) = X(t)/R \in [0, 2\pi)$ and N reveals the N-fold rotational symmetry of the ICDW ring.

Suppose that an ICDW ring is threaded by an Aharonov-Bohm flux Φ. Then, the effective Lagrangian for the phase of a rigid-body ICDW ring is given by [12, 16]

$$L(\dot{\xi}, \xi') = \int_0^{2\pi R} \mathrm{d}x \left[\frac{\hbar}{4\pi v'}\dot{\xi}^2 - \frac{c_0^2}{2}\xi'^2 + \frac{e}{\pi}A\dot{\xi}\right] \tag{4}$$

where $\dot{\xi} = \partial\xi/\partial t$, $\xi' = \partial\xi/\partial x$, $v' = c_0^2/v_\mathrm{F}$, c_0 is the phase velocity, v_F is the Fermi velocity in the conductor, and $\hbar$ is the reduced Planck constant. $A = \Phi/2\pi R$ is the magnitude of the vector potential along the ring perimeter. This vector potential cannot be gauged away continuously because the ICDW ring is not simply connected. ξ does not depend on position, hence $\dot{\xi}$ does not depend on position either and $\xi' = 0$. Therefore, equation (3) combined with equation (4) gives

$$L(N\dot{\phi}) = \frac{\hbar R}{2v'} N^2 \dot{\phi}^2 + \alpha\hbar N\dot{\phi} \tag{5}$$

which defines the canonical angular momentum and Hamiltonian

$$\pi_{N\phi} = \frac{\partial L}{\partial(N\partial\phi/\partial t)} = \frac{R\hbar}{v'}N\dot{\phi} + \alpha\hbar, \tag{6}$$

$$H = \pi_{N\phi}N\dot{\phi} - L(N\dot{\phi}) = \frac{v'}{2R\hbar}\left(\pi_{N\phi} - \alpha\hbar\right)^2 \tag{7}$$

where $\alpha = \Phi/\Phi_0$ and $\Phi_0 = 2\pi\hbar/2e$ is the magnetic flux quantum. Note that the above definition of canonical angular momentum $\pi_{N\phi}$ is equivalent to the canonical momentum of the phase $\pi_\xi = \partial L(\dot{\xi}, \xi')/\partial\dot{\xi}$.

The ICDW ring system is quantized by transforming $N\phi$ and $\pi_{N\phi}$ to operators $\hat{N\phi}$ and $\hat{\pi}_{N\phi}$, which satisfy the commutation relation $[\hat{N\phi}, \hat{\pi}_{N\phi}] = i\hbar$. Note that the number of crests N is fixed by the Fermi wavelength k_F as in equation (2). $\hat{\pi}_{N\phi} = -i\hbar\partial/\partial N\dot{\phi}$ is the canonical angular momentum operator defined as the generator of rotational motion. The quantized Hamiltonian satisfies the time dependent Schrödinger equation

$$\begin{aligned} i\hbar\frac{\partial}{\partial t}\psi(N\phi, t) &= \frac{v'\hbar}{2R}\left(-i\frac{\partial}{\partial N\phi} - \alpha\right)^2 \psi(N\phi, t) \\ &= E\psi(N\phi, t). \end{aligned} \tag{8}$$

For $N = 1$, this is the Schrödinger equation of a particle on a ring threaded by a magnetic flux given in Wilczek's "ring particle model"[1]. For $N > 1$, the eigenstates of the Schrödinger equation are

$$\begin{aligned} \psi_l(N\phi, t) &= \langle N\phi, t | l, E_l\rangle = \mathcal{A}\exp(ilN\phi - iE_l t/\hbar) \\ E_l &= \frac{v'\hbar}{2R}(l - \alpha)^2 \end{aligned} \tag{9}$$

where $|N\phi\rangle$ is an orthonormal angular position state, $|l, E_l\rangle$ is the simultaneous eigenstate of angular momentum and energy and $\mathcal{A}$ is a normalization factor to be determined. The periodicity $\psi_l(N\phi, t) = \psi_l(N(\phi + \lambda/R), t)$ implies that the angular momentum expectation values are quantized. Their values are $\langle l|\hat{\pi}_{N\phi}|l\rangle = l\hbar$ where l is an integer. The internal periodicity $|N(\phi + \lambda/R)\rangle = |N\phi\rangle$ implies that the probability of observing an ICDW crest within interval $0 < R\phi \leq \lambda$ is unity. In other words, for any ϕ and $\phi' = \tilde{\phi}'_m + m\lambda/R$, where $0 < R\tilde{\phi}'_m \leq \lambda$ and m is an integer, the orthogonality condition is given by

$$\int_0^{\lambda/R} \langle N\phi | N\phi'\rangle \mathrm{d}\phi = \int_0^{\lambda/R} \langle N\phi | N\tilde{\phi}'_m\rangle \mathrm{d}\phi \tag{10}$$

$$= \int_0^{\lambda/R} \delta(\phi - \tilde{\phi}'_m)\mathrm{d}\phi = 1. \tag{11}$$

The condition

$$\int_0^{\lambda/R} |\psi_l(N\phi,t)|^2 R d\phi = 1 \tag{12}$$

gives $\mathcal{A} = \lambda^{-1/2}$. Therefore, the general wave function of a rigid-body ICDW ring is

$$\begin{aligned}\psi(N\phi,t) &= \sum_l c_l \psi_l(N\phi,t) \\ &= \sum_l \frac{c_l}{\sqrt{\lambda}} \exp[i(lN\phi - E_l t)/\hbar]\end{aligned} \tag{13}$$

where c_l is the probability amplitude for mode l.

ICDW AS QUANTUM SPACE CRYSTAL AND QUANTUM SPACE-TIME CRYSTAL

Now, we show that an ICDW ring forms a QSC when α is an integer and forms a QSTC otherwise. A CDW transition occurs due to electron-phonon interaction, so an ICDW ring is fundamentally a macroscopic $2k_{\mathrm{F}}$-phonon system. For rigid-body ICDW rings, we define the density operator

$$\hat{\varrho} = |\Psi\rangle\langle\Psi| = \sum_{l,l'} \varrho_{ll'} |l, E_l\rangle\langle l', E_{l'}|, \tag{14}$$

where $|\Psi\rangle = \sum_l c_l |l, E_l\rangle$, and the momentum representation is $\varrho_{ll'} \equiv \langle l, E_l|\hat{\varrho}|l', E_{l'}\rangle = c_l c_{l'}^*$. We consider the Canonical ensemble [17]

$$\hat{\varrho} = e^{-\beta \hat{H}} / \sum_l e^{-\beta E_l},$$

where $\beta = 1/k_B T$, k_B is the Boltzmann constant and T is the temperature. The off-diagonal components of $\varrho_{ll'}$ vanish. The diagonal components ϱ_{ll} are calculated using the transformation $l - \alpha = (l - l_0) + (l_0 - \alpha)$, where l_0 is an integer that minimizes $|l_0 - \alpha|$:

$$\varrho_{ll} = \frac{\exp(-\beta E_l)}{\sum_{l'} \exp(-\beta E_{l'})} \to \delta_{l,l_0} \text{ as } T \to 0. \tag{15}$$

δ_{l,l_0} is the Kronecker delta. We propose the following time-dependent density matrix:

$$\begin{aligned}\varrho(\phi,\phi',t,t') &\equiv \langle \phi, t|\hat{\rho}|\phi', t'\rangle \\ &= \sum_l \frac{\varrho_{ll}}{\lambda} \exp[il(\phi'-\phi) - iE_l(t'-t)/\hbar].\end{aligned} \tag{16}$$

If α is an integer, $l_0 = \alpha$ and the time dependent density matrix becomes

$$\varrho(\phi,\phi',t,t')|_{\alpha\in\mathbf{Z}} \to \frac{\exp[il_0(\phi'-\phi)]}{\lambda} \text{ as } T \to 0. \tag{17}$$

The density matrix does not vanish as $|\phi - \phi'| \to \infty$, $\forall\, \phi, \phi'$, which is exactly the definition of ODLRO.

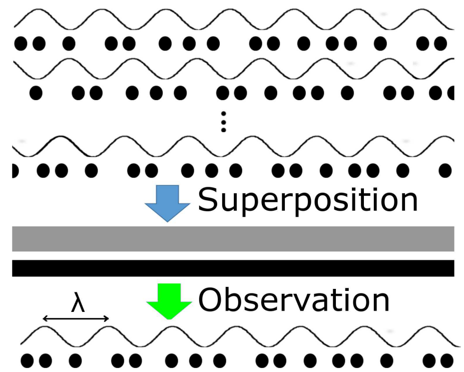

FIG. 1. The wave represents charge densities of an ICDW and the black circles represent atoms in a ring crystal. If α is an integer, the ground state of an ICDW rigid-body is described by ODLRO, i.e. the superposition of ICDWs with different phases. If the system is observed, the superposition is broken and the spontaneous breaking of the space translation symmetry becomes apparent.

Therefore, the ground state of an ICDW is an ODLRO spatial crystal (i.e. a QSC, see figure 1). If α is not an integer, $l_0 - \alpha \neq 0$ and the density matrix still does not vanish for $|\phi - \phi'| \to \infty$ and $|t - t'| \to \infty$. This implies that the ICDW ring exhibits ODLRO in both space and time. $\langle \hat{\pi}_{N\phi}\rangle = l\hbar$ is the expectation value of the *canonical* angular momentum operator, but the observable mechanical momentum is given by $\langle \hat{\pi}_{N\phi} - \alpha\hbar\rangle = (l-\alpha)\hbar$. In fact, from the definition of collective coordinate $X(t) = \xi(t)/2k_{\mathrm{F}}$, we can define the drift velocity of the ICDW as $v_{\mathrm{d}}^{\alpha} = \dot{\xi}(t)/2k_{\mathrm{F}}$, and using equations (5) and (6) we obtain

$$v_{\mathrm{d}}^{\alpha} = \frac{v'}{N} \sum_l \varrho_{ll}(l-\alpha). \tag{18}$$

So, the effective energy

$$U = \mathrm{tr}(\hat{\varrho}\hat{H}) = \sum_l \varrho_{ll} E_l \tag{19}$$

$$\to E_{l_0} = \frac{v'\hbar}{2R}(l_0-\alpha)^2 \text{ as } T \to 0 \tag{20}$$

and the current produced by the sliding ICDW ring [8]

$$\begin{aligned}j^{\alpha} = eN_e v_{\mathrm{d}}^{\alpha} &= \frac{ev'}{\pi R} \sum_l \varrho_{ll}(l-\alpha) \\ &\to \frac{ev'}{\pi R}(l_0-\alpha) \text{ as } T \to 0\end{aligned} \tag{21}$$

do not vanish for $\alpha \notin \mathbb{Z}$, where $N_e = 2k_{\mathrm{F}}/\pi$ is the number of conducting electrons per unit length. If α is an integer, the ground state of the ICDW is a stationary state with $v_{\mathrm{d}}^0 = 0$. However, if $\alpha \neq 0$, the ground state is the ICDW rotating with drift velocity $v_{\mathrm{d}}^{\alpha} \neq 0$. Therefore, there is a non-zero current for a finite ring even at $T = 0$. Because an ICDW is a QSC in the ground state,

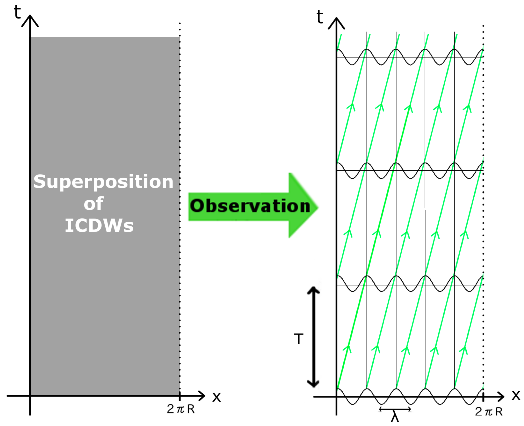

FIG. 2. Extension of figure 1 in the time domain. The unobserved system is an infinite superposition of lattices in space and time that does not break the space-time translation symmetry. Once the system is observed, the superposition is broken and the spontaneous breaking of the space-time translation symmetry becomes apparent. The arrows indicate the world lines of the ICDW crests.

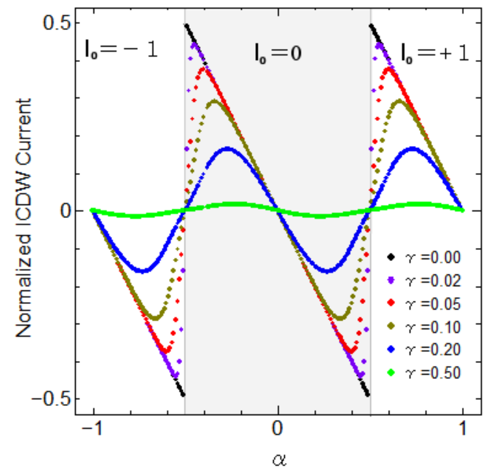

FIG. 3. There is a non-zero ICDW current even at $T = 0$ depending on the value of α, provided that the ICDW is formed on a perfect defect-free ring crystal. The temperature dependence is shown using a dimensionless parameter $\gamma = 2Rk_BT/v'\hbar$. The current is observable at finite temperature (millikelvin order).

a rotating ICDW ring would form a QSTC (see figure 2). In other words, the ground state of an unobserved ICDW ring is a superposition of lattices in space and time. The superposition is spatially and temporally uniform like a supercurrent state, hence the ground state of an unobserved ICDW ring does not break space-time translation symmetry although it is rotating. This is the *true* ground state of an ICDW ring. However, when the position of an ICDW crest is observed (e.g. observe with a photon or by using a probe that detects the charge modulation), the superposition is broken and the space-time translation symmetry becomes apparent. This is the *effective* ground state of an ICDW ring.

DISCUSSION

We first discuss how our model can be tested experimentally. Ring-shaped crystals (such as TaS_3 ring crystals) and ring-shaped CDWs have been produced and the coupling between CDW rings and vector potentials was verified by the Hokkaido group [18–20]. Therefore, our model can be tested provided that rings with almost no defects can be produced. Real systems can never achieve $T = 0$, so the temperature dependence is simulated using the dimensionless parameter $\gamma = 2Rk_BT/\hbar v'$. For a ring with diameter $2R = 10\mu$m, $m^*/m \sim 1000$ and $v_{\mathrm{F}} \sim 10^6$m/s, we have $T \sim \gamma \times 10$mK, and the qualitative behavior of ICDW current will be observed below $\sim$ 1mK (or $\gamma = 0.1$). The sliding of the ICDW ring implies that the local charge density oscillates with frequency $\omega = Nv_{\mathrm{d}}^{\alpha}/R \approx v'(l_0 - \alpha)/R \sim (l_0 - \alpha) \times 10^8$Hz. Therefore, the phason mode of an ICDW ring can be observed using femtosecond spectroscopy [21] or by resonating a cantilever to observe a behavior similar to Shapiro step. It is interesting that the ICDW current *emerges* as the temperature decreases.

Next, we discuss how our model avoids criticisms of QTCs. The main argument of Bruno's "no go theorem"[5] is that a quantum system cannot spontaneously break time translation symmetry because the energy of a rotating system is always larger than that of a system at rest. This theorem disproves the existence of QTCs provided that the energy spectrum of the system contains a stationary state [22]. However, our model is described by a single-particle energy spectrum that does not contain a stationary state. For $R \to \infty$, the ground state asymptotically approaches the stationary state, but a ring with a finite radius can rotate in the ground state. Bruno also argued that spontaneously rotating ground states cannot exist because of radiation [23]. However, this argument applies to an observed QTC whose superposition is broken. An unobserved QTC, including our model, is not subject to such arguments [24].

Another criticism is made by Watanabe and Oshikawa [6]. They defined time crystals in terms of correlation functions and proved a certain inequality, which shows that these correlation functions must be time independent. More precisely, the density matrix that we used is a kind of correlation function, and we can write Wanatabe and Oshikawa's inequality using the time dependent den-

sity matrix:

$$|\varrho(x,x',t,t') - \varrho(x,x',t',t')| = \left|\sum_l \frac{2\varrho_{ll}}{\lambda}\sin\left[\frac{E_l}{\hbar}(t'-t)\right]\right| \le \sum_l \frac{2|\varrho_{ll}|}{\lambda}\frac{E_l}{\hbar}(t'-t) = \sum_l \frac{|\varrho_{ll}|k_{\mathrm{F}}v'}{\pi R}(l-\alpha)^2(t'-t).$$

Watanabe and Oshikawa assumed that $(t-t')/R \to 0$ as $R\to\infty$ and that the dynamical state is asymptotically equivalent to the stationary state. However, for our model, $|t'-t| \sim \lambda/v_{\mathrm{d}} \sim R$ and Watanabe and Oshikawa's argument fails. This result originates in the internal periodicity of the system, which Watanabe and Oshikawa did not consider.

We expect that many other incommensurate systems such as incommensurate spin density waves [8], incommensurate mass density waves [25–27], or possibly some dielectrics that exhibit incommensurate phases [28], may be used to model and test QTCs.

CONCLUSION

In this paper we first showed that the rigid-body model of an ICDW ring is described by a macroscopic single-particle wave function. We also showed, using density matrices, that an ICDW ring is a QSC. The rotation of an ICDW ring is described by a gapless phason mode, so the ICDW ring can rotate in the ground state by coupling with an electromagnetic vector potential and form a QTC. A rotating ICDW ring still exhibits ODLRO in space, hence it is a QSTC that combines the concept of QSCs and QTCs. We also proposed that the rotation can be observed from the oscillation of the local charge density, which is possible at finite temperatures. Our model agrees with Bruno's "no-go theorem" [5] if the ICDW ring has an infinite radius, but an ICDW ring with a finite radius can rotate in the ground state. Moreover, we showed that another "no-go theorem" given by Watanabe and Oshikawa fails for systems with internal periodicity as in our model. Therefore, our result implies that QTCs can exist. QSCs and QTCs are two important predictions. They will be confirmed simultaneously if our model is verified experimentally.

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